

REMARKS

Reconsideration is respectfully requested of the rejections under 35 U.S.C. §§112 and 103

Rejections under 35 U.S.C. §112

In view of the instant amendments, reconsideration is respectfully requested of the rejection of claims 13-22, 38, 43, 48-99 and 165-199 under 35 U.S.C. §112. As amended, it is believed that claims 13-22, and 155-199 fully meet the concerns expressed by the Examiner in the Office action, and fully satisfy §112, first paragraph, in particularly pointing out and distinctly claiming the subject which Applicants regard as their invention. It is further respectfully submitted that original claims 38, 43 and 48-99 were in compliance with §112, first paragraph, so that no amendment was or is required.

Each of the rejected claims is directed to a process for the preparation of N-(phosphonomethyl)glycine or a salt thereof comprising contacting N-(phosphonomethyl)iminodiacetic acid or a salt thereof with an oxidation catalyst in the presence of oxygen. The catalyst comprises a carbon support having a noble metal, carbon, and oxygen at the surface of the carbon support.

Each of the rejected claims further specifies a minimum ratio of carbon to oxygen atoms and/or maximum ratio of oxygen to noble metal atoms at the surface of the carbon support. Claims 13 and 155 have been amended to specify the conditions under which the C/O and O/noble metal ratios are to be measured, i.e., the characterizing ratio is measured after the catalyst:

"is heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before the catalyst is exposed to an oxidant following the heating in the hydrogen atmosphere.."

Heating for one hour under such conditions substantially removes molecular oxygen and allows proper measurement of the ratio of carbon and/or noble metal atoms to atoms of oxygen bound to the surface. Note also that the claims specify determination of the C/O ratio by x-ray photoelectron spectroscopy, which is conventionally conducted under high vacuum conditions which further serve to remove any residual molecular oxygen and prevent oxygen re-adsorption.

In response to the Examiner's inquiry at page 2 of the Office action, oxygen that is contained in "oxygen-containing functional groups is "covalently bonded," i.e., the latter description encompasses the former. Accordingly, there is no conflict in these terms. Also, the Examiner is correct in understanding that "in the presence of oxygen" comprises molecular oxygen in the reaction atmosphere, and that "oxygen at the surface" and "in the presence of oxygen" are not being used interchangeably.

When the claims were reviewed in light of the §112 rejection, it was recognized that original claims 13, 155 and 165 were more restricted in scope than had been intended, i.e., they could have been read as requiring that the specified ratio of carbon atoms to surface oxygen atoms prevails even in the presence of molecular oxygen during the claimed oxidation reaction, a condition not practical to measure and perhaps unlikely to be achieved. It is important, indeed critical, to understand that Applicants have provided a noble metal on carbon catalyst of superior performance properties which can be identified by one more of the various characterizing criteria of the claims, e.g., various claims are satisfied if either:

1. the catalyst yields no more than about 1.2 mmole of carbon monoxide per gram of catalyst when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before

being exposed to an oxidant following the heating in the hydrogen atmosphere, is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes in a helium atmosphere, as specified in claims 1-11, 75-78 and 150-199;

2. the C/O atomic ratio is at least about 20:1 at the surface of the carbon support after exposure of the noble metal on carbon catalyst to a hydrogen atmosphere at 500°C for an hour and before the catalyst is exposed to an oxidant following heating in a hydrogen atmosphere, as specified in claims 13-22, 66-78 and 155-199;

It is possible, indeed likely, that the bound oxygen content of the catalyst surface increases upon exposure to molecular oxygen during the course of the claimed oxidation reaction. However, if the catalyst meets the characterizing criteria specified in the claims, it provides advantageous performance properties despite exposure to a strongly oxidizing atmosphere during the conduct of the process as claimed.

It should be understood that the instant amendments to the claims are not narrowing amendments. On the contrary, they are effectively broadening amendments since claims 13, 155 and 165 as originally presented might have been construed as requiring that the specified C/O ratios be met during the course of the reaction which, as noted, they may not.

Claim 1 has also been amended in a manner comparable to the amendment of claim 13, and for the same purpose.

Common Ownership

Applicants respectfully acknowledge their obligation to identify any difference in ownership among the invention(s) defined by the various claims, and to identify the earlier of the inventions of any two claims which were not commonly owned at the

time the later invention was made. This matter has been reviewed, and Applicants can report that all the inventors in the instant application were employees of the assignee hereof at the time the invention(s) of the various claims were conceived and reduced to practice.

In the accompanying Information Disclosure Statement, Applicants have identified information received from a third party regarding a possible continuous process for the preparation of N-(phosphonomethyl)glycine using a carbon catalyst which did not include any noble metal. Assuming *arguendo* that this information qualifies as §102(f) prior art and, if so, that the information is prior art for purposes of §103, it is respectfully submitted that it does not render obvious the invention as claimed herein.

Rejections under 35 U.S.C. §103

Reconsideration is respectfully requested of the rejection of claims 1 to 199 under 35 U.S.C. §103 as unpatentable over Franz US 4,147,719 in view of Hess US 3,340,097, Hershman US 3,969,398, Coloma et al. J. Catal. 1995 and Ebner et al 5,627,125.

The Claimed Invention

All claims of the instant application are directed to a process for the oxidation of an N-(phosphonomethyl)iminodiacetic acid substrate to produce N-(phosphonomethyl)glycine ("glyphosate") using a noble metal on carbon catalyst. The noble metal on carbon catalyst is effective both for the oxidation of the substrate and for oxidation of C₁ by-products of the reaction, viz., formaldehyde and formic acid.

Deeply Reduced Catalyst

Claims 1 to 52, 66-99, and 150-199 require the use of a "deeply reduced" noble metal on carbon catalyst which is

effective for the oxidation of both the substrate and the C₁ by-products. In accordance with the invention, Applicants have discovered that, by use of such deeply reduced catalysts which meet the various characterizing standards respectively set forth in the claims, N-(phosphonomethyl)iminodiacetic acid can be substantially quantitatively oxidized to N-(phosphonomethyl)glycine, and the aforesaid C₁ by-products can also be oxidized. Significantly, Applicants have further discovered that such oxidation reactions can be accomplished without the excessive loss of noble metal from a carbon support, as is otherwise typically encountered in acidic aqueous oxidation media.

More particularly, claim 1 is directed to a process for the oxidation of N-(phosphonomethyl)iminodiacetic acid or a salt thereof to glyphosate or a salt thereof using a noble metal on carbon catalyst that is characterized as yielding no more than about 1.2 mmole of carbon monoxide per gram of catalyst when a dry sample of the catalyst in a helium atmosphere is heated from about 20° to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes. To provide a catalyst which gives such a low CO desorption yield, the catalyst is "deeply reduced" in accordance with the methods described in the specification of the application.

As discussed hereinabove, claims 13-22, 66-78 and 155-199 define a deeply reduced catalyst in somewhat different terms, i.e., with respect to the C/O ratio at the surface of the carbon support. Claims 23-32 and 79-99 characterize the catalyst with respect to the C/O ratio in a surface layer having a thickness of about 50 Å as measured inwardly from the surface.

Claim 33-52 and 165-199 define a deeply reduced catalyst with reference to the methods for its preparation. Thus, claims 33-48 call for depositing a noble metal at a surface of a carbon support, and then heating the surface at a temperature of at least about 400°C. Claims 38, 43, 49-52 and 165-199 all require

that, before noble metal deposition, the carbon support has carbon and oxygen at the surface of the carbon support in amounts such that the ratio of carbon atoms to oxygen atoms at the surface is at least 20:1, again as measured by x-ray photoelectron spectroscopy.

Continuous Oxidation

Applicants have further discovered that, by use of a noble metal catalyst, barriers to continuous catalytic oxidation can be overcome; and in particular that the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate can be conducted in a continuous reaction mode. This enables major advantages to be realized with respect to capital investment, productivity, energy recovery, emission control, etc. Thus, Claims 2, 14, 24, 34, 50, 54, 80 and 100 to 199 are directed to a continuous process for the preparation of glyphosate by catalytic oxidation of N-(phosphonomethyl)-iminodiacetic acid. Claims 100-199 specifically require the use of a particulate noble metal on carbon catalyst in a stirred tank reactor or a continuous reactor system.

Applicants have further discovered that continuous reaction can be feasibly and economically carried out in the relatively back-mixed conditions prevailing in continuous stirred tank reactors. Claims 100-199 are all directed to a continuous process as conducted in a continuous reaction system which includes a CSTR.

Promoted Catalysts

Claims 53-99 and 144-199 call for the use of a noble metal on carbon catalyst that further comprises a promoter. Promoters have been found to significantly enhance the effectiveness of the catalyst for oxidation of substrates such as N-(phosphonomethyl)iminodiacetic acid, and/or by-products such as formaldehyde and formic acid, in an acidic aqueous medium.

The use of a promoter further enhances the effectiveness of the continuous process as defined in claims 54 and 144-199.

Patentability over the Prior Art

It is respectfully submitted that none of the references relied on in the action, whether considered individually or in combination, would have rendered the claimed invention obvious to one of ordinary skill in the art.

Franz US 4,147,719

Franz is illustrative of the state of the art prior to the instant invention. As noted by the Examiner, Franz describes the preparation of glyphosate by oxidation of an aqueous solution of N-(phosphonomethyl)iminodiacetic acid with a heterogeneous catalyst comprising Pt or Rh on carbon.

However, Franz fails to teach or suggest a process for preparation of glyphosate using a deeply reduced catalyst of the type called for in claims 1-52, 66-99, or 150-199. Although the catalyst as disclosed by Franz is capable of oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate, and is also effective for the oxidation of C₁ by-products such as formaldehyde, the process as described by Franz has never been commercialized, primarily because of evidence that Pt would be rapidly lost from the catalyst surface under the acidic oxidation conditions prevailing in the conversion of N-(phosphonomethyl)iminodiacetic acid to glyphosate. Franz betrays no recognition of the problem of noble metal dissolution in the aqueous reaction medium, and offers no method, stratagem or technique to deal with this problem. In practical reality, metal loss from a catalyst such as that disclosed by Franz is too excessive for the Franz process to be competitive with known commercial processes that use carbon catalysts containing no catalytically effective amount of noble metal.

Franz entirely fails to disclose or suggest a catalyst having the limited CO desorption characteristic of the catalyst that is used in the process of claim 1-11, 75-78 or 150-199; or the high C/O ratios of the catalyst of claims 13-32, 66-99, or 155-199. Nor does Franz provide any suggestion of preparing the catalyst by a method in which the C/O ratio at the surface is greater than 20:1 at the time of metal deposition, as specified in claims 38, 43, 48, 49-52 and 165-199; or by the method in which the catalyst is heated to 400°C as required by claims 33-48.

Much less does Franz recognize that deeply reduced catalysts offer the key to achieving the benefit of noble metal on carbon catalysts without the excessive metal loss that ordinarily attends the use of these catalysts in an aqueous acidic oxidation medium.

As the Examiner has further recognized, Franz also fails to teach a continuous process for the oxidation of N-(phosphonomethyl)-iminodiacetic acid to glyphosate, as is required by claims 2, 14, 24, 34, 50, 54, 80 and 100-199. Applicants respectfully but strenuously take issue with the Examiner's reliance on MPEP 2144.04, section V-E as establishing that "going from a batch process to a continuous process is an obvious modification." As a matter of practical industrial experience, the conversion of a batch to a continuous process commonly presents difficult, complex and often insuperable technical barriers. This is especially so in the case of processes which comprise chemical reaction. Continuous processes offer certain potential advantages, so that conversion of a process from batch to continuous may be commonly viewed as desirable, at least superficially. But there are pitfalls in continuous operation; and the fact that a high proportion of manufacturing processes, especially in the chemical industry, remain as batch operations is telling testimony to the unobviousness of converting to continuous.

In processes comprising a chemical reaction step, problems of reaction kinetics, competing reactions, secondary reactions, heat transfer, mass transfer, and process control often render continuous operation difficult, impracticable, or impossible, especially in reactions that are exothermic. Note that MPEP 2144.04 does not establish any categorical rule that it is obvious to make a batch process continuous; nor could it since any such "negative maxims" of unpatentability were abolished by the enactment of the obviousness standard of 35 U.S.C. §103.

Whether a continuous process is obvious or not obvious from the prior art depends, of course, on the nature of the process and the prior art. Obviousness *vel non* is to be determined under the *Graham* standards, not by negative rules that may be derived from early case law, especially - as here - where the adverse authority is readily distinguishable. The MPEP cites *in re Dilnot*, wherein the invention was directed to a process for the preparation of a cellular cementitious structure. The claim at issue called for:

"The method of producing a cellular cementitious structure consisting of the steps of, preparing a slurry of cementitious material, generating a stable air foam, continuously introducing said foam into said slurry, and intimately mixing said foam with said slurry as said foam is introduced to produce a substantially uniform dispersion of said foam and in said slurry, introducing said mixed foam and slurry into a form of desired configuration, and allowing said mixture to set and harden."

It will be noted that the only step required to be continuous is the physical step of introducing the foam into the slurry. The only chemical reaction step of the process is "allowing said mixture to set and harden" after it is introduced into the form,

a step which the Dilnot claim allows, if it does not actually require, to be conducted in batch mode.

Accordingly, the MPEP and the *Dilnot* case provide no authority for the rejection of the claims of the instant application which call for conducting the catalytic oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate on a continuous basis. Unlike the physical mixing step of *Dilnot*, the claimed continuous reaction involves kinetic limitations, exothermic heat effects, heat transfer requirements, actual or potential mass transfer limitations, potential oxidation of desired reaction product, and consequent process control issues. Moreover, the process produces by-product C₁ compounds which are reactive with the glyphosate product to produce undesired glyphosate derivatives that reduce yields, and potentially affect product quality and/or complicate product recovery. Upon hearing any proposal to conduct the exothermic reaction of this invention continuously in a back mixed condition, one skilled in the art might expect that kinetic effects would be unfavorable, that high steady state concentrations of glyphosate would result in over-oxidation with increased formation of by-product aminomethylphosphonic acid. High steady state glyphosate concentration would also have been expected to result in increased formation of undesired derivatives such as N-methyl glyphosate, and that the latter effect would be aggravated by a possibly high steady state concentration of C₁ by-products in the reaction zone. On the other hand, if the process were operated in a plug flow reactor, problems of heat transfer, mass transfer, catalyst configuration and multi-phase flow would create other potentially major complications. These various and multiple problems render design and operation of a continuous catalytic oxidation step at least potentially far more complex and difficult than the simple air/foam mixing step of *Dilnot's* process.

Nonetheless, in accordance with the present invention, Applicants have overcome problems of the type noted hereinabove, and thereby developed a practical and effective continuous process for the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate. More particularly, Applicants have discovered that continuous oxidation of N-(phosphonomethyl)-iminodiacetic acid to glyphosate can be conducted using a noble metal on carbon catalyst, which is effective both for the oxidation of the substrate to glyphosate and oxidation of the by-product formaldehyde and formic acid to carbon dioxide and water.

As defined in claims 100-199, Applicants have further discovered that a continuous catalytic oxidation process can be effectively conducted in a continuous reactor system comprising a stirred tank reactor in which the substrate is contacted with an oxygen source and a particulate catalyst comprising a carbon support having a noble metal at the surface of the carbon support. Nothing in the Franz reference suggests that the oxidation of N-(phosphonomethyl)-iminodiacetic acid to glyphosate can feasibly be conducted in such a continuous reaction system. As the Examiner has acknowledged, the Franz reaction system is entirely batch.

The process of claims 100-199 has been enabled by Applicants' recognition of the unique capabilities of noble metal on carbon catalysts in the production of glyphosate. Further in accordance with the invention, Applicants have recognized not only that noble metal on carbon catalysts are effective for oxidation of both substrate and by-products, but also that the kinetics of the reaction allow substantially quantitative conversion to glyphosate within a commercially economical residence time in reaction systems comprising back mixed reactors; and further that such result can be achieved without undue formation of glyphosate derivatives such as N-methyl glyphosate or aminomethylphosphonic acid. Franz entirely fails to suggest such a system. Much less does Franz suggest the

continuous process as defined in claims 150-199 wherein a deeply reduced particulate noble metal on carbon catalyst is used in the continuous oxidation of N-(phosphonomethyl)iminodiacetic acid in a reactor system comprising a stirred tank reactor.

As acknowledged in the Office action, Franz further fails to disclose or suggest the oxidation of N-(phosphonomethyl)-iminodiacetic acid to glyphosate in the presence of a noble metal on carbon catalyst that further comprises a promoter, as defined, for example in claims 53-99 and 144-199. Franz further fails to characterize the catalyst by surface area, particle size, etc.

While even earlier Monsanto patent references briefly describe a continuous process in which N-(phosphonomethyl)-iminodiacetic acid is oxidized to glyphosate in a flow reactor comprising a fixed bed of carbon, none of the latter references describe the use of a noble metal on carbon catalyst. Moreover, none of the flow reactor systems of these earlier patents has been demonstrated to offer a commercially feasible alternative for the manufacture of glyphosate, as has the process of the present invention.

It is therefore respectfully submitted that the instantly claimed invention is not rendered obvious by the teachings of Franz, or of any other prior art references relating to the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate.

Hess et al US 3,340,097

The Office action relies on the Hess reference as teaching that metals other than noble metals, in combination or as alloys, have a promotional effect on the catalytic activity of platinum.

It is respectfully noted that Hess has no relation whatsoever to the catalytic oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate. On the contrary, Hess describes a fuel cell operation. For use in a fuel cell Hess describes electrodes having a surface codeposit of Pt, Ru and Sn. The only reaction disclosed is the

electrochemical oxidation of methanol in sulfuric acid or potassium hydroxide solution.

The Office action identifies no basis or motivation to combine Hess with Franz. The two are in disparate arts and neither contains any teaching from which one skilled in the art would be led to combine it with the other. Franz oxidizes a substrate with molecular oxygen in the presence of a noble metal on carbon catalyst. Hess does not contact his methanol substrate with molecular oxygen but instead causes release of electrons to the anode of a fuel cell. Thus, even by hindsight, it is difficult to say how Hess could be construed to offer guidance in the preparation of catalysts for use in the process of Franz.

Hess suggests that promoters improve the performance of fuel cell electrodes, but the electrochemical oxidation of methanol is much too far removed from the catalytic oxidation of N-(phosphonomethyl)iminodiacetic acid for Hess to have any meaning in the context of Franz.

Moreover, Hess further fails to teach or suggest the use of a catalyst or electrode prepared by deeply reducing the carbon support, and utterly fails to suggest catalysts that can be characterized by the CO desorption or C/O surface ratio characteristics of the deeply reduced catalysts used in the process of claims 1 to 99 and 150-199. Nor does Hess contain any suggestion of the processes by which the catalysts of claims 33-48 or 49-52 are prepared.

Hess has nothing whatsoever to say about any continuous process for the manufacture of any useful oxidation product. The Hess disclosure is primarily concentrated on the electrolytic processes for preparing the electrodes of his invention, not their use in generating energy by oxidation of a substrate such as methanol. However, to the extent that the methanol oxidation processes of the fuel cell can be understood from the Hess description, they would appear to operate in a batch mode.

In any case, Applicants do not understand the Examiner to be attempting to apply the Hess reference to the continuous process of claims 100-199.

Thus, it is respectfully submitted that claims 1-199 distinguish patentably over the combination of Franz and Hess under 35 U.S.C. §103.

Hershman US 3,969,398

The Office action cites Hershman as describing the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate using an activated carbon catalyst having a B.E.T. surface area of 100 to 2000 m²/g and a particle size of about 20 to 325 mesh, as indeed it does.

However, Hershman describes the use of a carbon catalyst only, not a noble metal on carbon catalyst as is required in all of claims 1 to 199. Thus, Hershman's catalyst is relatively ineffective for the oxidation of formic acid or formaldehyde, and is consequently unsuited for continuous oxidation of N-(phosphonomethyl)iminodiacetic acid in a continuous stirred tank reactor ("CSTR") as required by claims 2, 14, 24, 34, 50, 54, 80 and 100-199. For if continuous oxidation were conducted in the presence of the high concentrations of unreacted C₁ by-products that would be present under the terminal conditions prevailing in a CSTR, as would be the case where Hershman's carbon catalyst is used, one skilled in the art would have expected unacceptable contamination with N-methyl glyphosate and possibly other glyphosate derivatives. Although carbon catalysts of the type described in Hershman and other Monsanto patents (e.g., Chou 4,696,772) were the state of the art prior to the present invention, they have not to Applicants' knowledge been used commercially in any continuous reaction system.

The Hershman reference further fails to teach or suggest a deeply reduced noble metal on carbon catalyst meeting the CO desorption or C/O ratio criteria of claims 1-32, 66-99 and 150-

199. Hershman indeed describes activating a carbon catalyst by heating it alone at 800° to 900°C in the presence of carbon dioxide or steam. However, there is no suggestion whatsoever in Hershman that such heating results in the preparation of even a carbon catalyst meeting the instantly claimed CO desorption and C/O ratios, nor that such parameters could have any relevance to the performance of the catalyst in the process of Hershman. Indeed there is no evidence of record that such parameters could be expected to have any significance in such context. Applicants have discovered deep reduction as a novel way to prevent dissolution of noble metal from the catalyst under the aqueous acidic conditions of the oxidation reaction. Since Hershman did not use noble metal, the issue of metal loss simply did not arise.

In any event, the combination of Hershman and Franz does not lead to either a deeply reduced catalyst having a CO desorption less than 1.2 mmols under conditions defined in claims 1-11, 75-78 and 150-199, or having a C/O ratio of at least 20:1 under the conditions of claim 13-32, 66-99 or 155-199. Nor does Hershman describe heating a noble metal on carbon catalyst at greater than 400°C, as called for in claims 33-48, nor reducing the carbon surface to provide a C/O ratio greater than 20:1 before noble metal deposition as specified in claim 38, 43, 48-52 and 165-199.

Nor does the combination of Franz and Hershman teach or suggest the continuous process of claims 2, 14, 24, 34, 50, 54, 80 or 100 to 199. Both Franz and Hershman describe batch reactions.

Combining Franz and Hershman with Hess still fails to yield a process for oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate that uses a deeply reduced noble metal on carbon catalyst; or a process which in which N-(phosphonomethyl)-iminodiacetic acid is oxidized to glyphosate in a reaction system comprising a CSTR.

Accordingly, it is respectfully submitted that claims 1-199 distinguish patentably over the combination of Franz, Hess and Hershman under 35 U.S.C. §103.

Coloma, J. Catal., 1995

Although Coloma et al. describe the preparation of Pt/C catalysts, the only use of such catalysts which the reference describes is the hydrogenation of benzene to cyclohexane. Manifestly, one skilled in the art would not be led to consult Coloma for teachings relevant to the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate, or for dealing with the problem of noble metal dissolution in an aqueous acidic oxidation environment.

Coloma et al. do not say whether they conducted benzene hydrogenations in a batch mode or continuous mode. In any event, there is no teaching in Coloma which could lead to a continuous process for the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate.

For the value it may have in the context of the catalytic hydrogenation of unsaturated hydrocarbons, Coloma et al. do describe treatment of a Pt/C catalyst surface at 350° to 500°C in hydrogen, and relate this treatment to metal dispersion as measured by H₂ and CO₂ adsorption. The reference also states that oxygen surface complexes may influence "other kinds of centers on the carbon surface;" and that the strength of the so-called π sites, that can act as anchoring sites for Pt, is diminished by the presence of oxygen complexes, due to electron withdrawing effect exerted by the oxygen (p. 298). However, this discussion is not referenced to any maximum CO desorption or minimum C/O ratio; and in any case does not relate to any oxidation reaction, especially not to the problem of metal loss under aqueous acidic oxidation conditions.

The Office action concludes that, "[t]he skilled artisan would be motivated to heat the carbon surface, before and/or

after the dispersion of the metal catalyst, in order maximize the activity of the carbon surface." It is respectfully submitted that this misses the point. The main object of the instant invention is not mere carbon activation. Aside from whatever carbon activation may incidentally be achieved, Applicants have overcome the severe metal dissolution problems which had precluded the use of noble metal on carbon catalysts in aqueous acidic oxidation environments such as the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate. Nothing in any of the references suggests any such effect, or the means by which it is achieved.

Ebner US 5,627,125

Ebner is apparently relied on only with reference to claims that call for a sacrificial reducing agent in the process, the action asserting that it would be obvious to keep the catalyst in a "maximally reduced state," citing col. 7, lines 34-48.

This part of the Office action seems to reflect some confusion, since the cited passage relates to the deposition of copper onto a Pt/C substrate, not deposition of Pt onto the carbon. Ebner is in any event directed to a dehydrogenation reaction, not a reaction of a substrate and an oxidant. Such dehydrogenation reactions are copper-catalyzed, and the only disclosed function of the Pt/C is to serve as a support for the copper so copper is not lost in the reaction. Ebner does not describe or suggest any relationship between the oxidation state of the carbon and any loss of noble metal.

Moreover, the dehydrogenation reactions conducted by Ebner are run under a reducing atmosphere, i.e., with the reaction mixture under hydrogen pressure resulting from the generation of hydrogen in the reaction. Thus, Ebner would not be seen by one skilled in the art as having any relevance to the catalytic oxidation reaction of the instant invention.

In any event, since Ebner is cited only against dependent claims of the application, patentability of these claims is adequately established by the patentability of the claims on which they depend. It is, therefore, respectfully submitted that claims 1-199 distinguish patentably over any combination of Franz, Hess, Hershman, Coloma and Ebner.

Promoted Catalysts

With respect to claims 53-99 and 144-199, Applicants have discovered that the oxidation processes of the invention are enhanced where the catalyst comprises one or more promoters such as Fe, Sn, Bi, Ti, etc. As noted above, and as the Examiner has acknowledged, neither Franz, Hershman nor Coloma contains any suggestion of the presence of a promoter. The only reference relied on as suggesting the use of a promoter is Hess US 3,340,097 which does not relate to any catalyst for oxidation of a substrate by reaction with molecular oxygen, but instead is directed only to electrodes for fuel cells. For the reasons discussed above, Hess is completely unrelated to Franz or Hershman or the art to which the present invention is directed. No basis has been suggested for combining Hess with any reference in the field of Applicants' invention. On this further ground it respectfully submitted that claims 53-99 and 144-199 calling for a promoter are patentable over the cited art under 35 U.S.C. §103.

Further Comments of the Office Action

The Examiner contends that, in the absence of evidence that the CO desorption and C/O ratios features of the claims are achieved only through the use of a specific support or treatment, "it must be concluded that this characteristic would be part of carbon-supported noble metal catalysts prepared by pre-heating the carbon material."

It is respectfully noted that the Examiner has provided no evidence to support the stated conclusion. The only two references relied on which describe noble metal on carbon catalysts are Franz and Coloma. Franz describes preparation of a Pt/C catalyst prepared by depositing Pt on a commercially available activated carbon support. The text expressly mentions the activated carbon of Hershman US 3,969,398. However, Franz describes no treatment of the carbon surface either before or after deposition of the platinum. It is therefore unlikely that the catalyst of Franz would exhibit a CO desorption of less than 1.2 mmoles or a C/O surface ratio of greater than 20:1 after exposure to hydrogen at 500°C for only one hour.

It is respectfully noted that a finding of inherency cannot be based on mere assumptions by the Examiner; *in re Rijkaert*, 28 USPQ2d 1955 (Fed. Cir. 1994). But even if it were assumed that the methods of Coloma or Hershman could achieve the claimed surface characteristics of the carbon, neither Franz, Hershman nor Coloma contain any teaching to such effect. Thus, there is no basis for combining Hershman or Coloma with Franz to reject any of the instant claims under 35 U.S.C. §103. "Obviousness cannot be predicated on what is unknown;" *in re Spormann*, 150 USPQ 449, 452 (CCPA 1966).

The references particularly fail to provide motivation for one skilled in the art to combine the oxidation process of Franz with the hydrogenation catalyst of Coloma. Certainly such motivation cannot be provided by the CO desorption or C/O surface ratio of the Coloma catalyst, which are not disclosed by Coloma and remain unknown.

There is manifestly no basis for concluding that the carbon of Hershman in fact possessed the deeply reduced carbon properties specified in the claims. Hershman says only that:

"Activation is usually achieved by heating to high temperatures (800°-900°C) with *steam or with carbon*

dioxide, which brings about a porous particle structure and increased specific surface area. in some cases hygroscopic substances, such as zinc chloride and/or phosphoric acid or sodium sulfate, are added prior to the destructive distillation or activation, to increase adsorptive capacity." (Emphasis supplied)

As described in the instant specification, Applicants' catalyst preparation preferably begins with such activated carbon (p. 20, lines 6-9) but does not end there. Applicants preferably conduct a further deoxygenation step before noble metal deposition, in which case steam and carbon dioxide, the atmospheres suggested by Hershman, are preferably avoided. As explained in the instant specification (p. 24, lines 24-28):

"as the temperature of the carbon support is cooled below 500°C, the presence of oxygen-containing gases such as steam or carbon dioxide may lead to re-formation of the surface oxides."

Thus, it appears likely that a carbon activated only by the Hershman method would be oxygenated to an extent exceeding the limitations of the instant claims. At any rate, given the minimal information and data on the activation process as described by Hershman, there is certainly no basis for concluding that Hershman carbon inherently possessed the CO desorption or C/O surface ratio parameters which characterize the catalyst used in the process of the present invention.

As described in the attached Third Supplemental Information Disclosure Statement, activated carbon produced in accordance with the Chou patent exhibits a CO desorption below 0.5 mmol/g, and a C/O surface ratio of about 30:1 under the conditions specified in claims 1, 13, 23 and 79. However, Chou relates solely to a carbon catalyst. The art of record contains no

suggestion of using the Chou catalyst as a substrate for a noble metal. Moreover, the surface oxide characteristics of Chou are not disclosed in any statutory prior art, and therefore cannot be deemed available to the hypothetical person of ordinary skill for purposes of analysis under 35 U.S.C. §103(a). Cf. *Kimberly-Clark v. Johnson & Johnson*, 223 U.S.P.Q. 603 (Fed. Cir. 1984).

The working examples of the instant application further demonstrate that commercially available activated carbons such as SA-30, and commercially available Pt on activated carbon catalysts such as that available from Aldrich, fail to satisfy the CO desorption and/or C/O surface ratio characteristics specified in the claims. See Table 1 (SA-30), Table 5 (5% Pt/SA-30; Aldrich 5% Pt/C), and Table 6 (Aldrich 5% Pt/C). The examples also demonstrate the general superiority of catalysts that are deeply reduced in accordance with the invention by processes that are effective to achieve the parameters set out in the claims.

Franz for his part fails to describe the handling of the carbon prior to platinum deposition, and fails to teach or suggest any reduction step following noble metal deposition. Thus, even if it were assumed *arguendo* that the Hershman activation method would reliably reduce bound oxygen to levels characterized by the CO desorption and C/O ratio parameters specified in the instant claims, there is no basis for concluding that the Franz catalyst would meet these parameters. Note that Applicants preferably reduce the catalyst after noble metal deposition. Claim 49 expressly requires a reduction step after deposition of a noble metal on a carbon support which has a greater than 20:1 C/O surface ratio before the deposition. In the case where there has been no reduction step prior to metal deposition, Applicants typically expose the catalyst to a reducing gas such as hydrogen for a lengthy period at an elevated temperature well above 500°C. See, for example, Tables 5 and 6. Applicants have selected such measures so that the requisite

degree of reduction can be consistently achieved. By comparison, the sketchy incidental disclosure of Franz and Hershman cannot conceivably be sufficient to establish inherent attainment of any quantitative degree of surface reduction. The passing allusion to activation falls far short of support for any conclusion that Franz or Hershman achieved the parameters severally specified in the instant claims. Inherency can be found only where attainment of a claimed feature is unavoidable, not where it is merely possible or even probable; *Continental Can v. Monsanto*, 20 USPQ2d 1746 (Fed. Cir. 1991); *ex parte Keith*, 154 USPQ 320 (PO Bd App 1966); *in re Oelrich*, 212 USPQ 323 (CCPA 1981); *in re Rijckaert*, *supra*. Neither Franz nor Hershman remotely meets this standard.

Dependent Claims

It is respectfully submitted that all dependent claims are patentable over the cited art on the same basis as the independent claims. However, further grounds of nonobviousness can be cited in support of all dependent claims. Prominent among the dependent claims are those discussed hereinbelow.

Series Reactors with Differing Oxygen Flow Regimes. In accordance with the invention, as defined in claim 117, it has been found that a continuous process for the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate can be effectively conducted in sequential reaction zones using differing oxygen flow regimes. Preferably, the reaction is conducted in two sequential CSTRs, with relatively reduced oxygen flow into the second reactor, as specified in claim 120. A relatively high oxygen flow is advantageously maintained in the first reactor wherein the reaction is essentially zero order to a very high conversion; and a lower oxygen flow is maintained in the second (or further) reactor(s). A high flow rate conduces to maximum conversion rate in the first reactor where progress of the reaction is typically mass transfer limited. In the second

reactor, a lower oxygen flow rate is preferred to prevent over-oxidation of the catalyst which may reduce its effectiveness for the oxidation of the C₁ by-products, formaldehyde and formic acid. Lower oxygen flow at low residual PMIDA concentration has been found effective for continued oxidation of formaldehyde without causing oxidation of glyphosate to undesired aminomethylphosphonic acid ("AMPA") by-product, though AMPA formation tends to be suppressed in any event under the preferred continuous process conditions using the preferred catalysts. Lower oxygen flow also helps to minimize dissolution of noble metal from the surface of the catalyst. On these further grounds it is respectfully submitted that claims 117 to 199 are patentable over the prior art under 35 U.S.C. §103.

Other Pending Dependent Claims. The Office action adopts the position that other claimed process features such as recovery and recycle of catalyst, temperature, pH, concentration of catalyst, etc. are result-effective variables that one skilled in the art would be expected to manipulate to advantage. It is respectfully submitted that the mere expression of expectation in this regard does not amount to a statutory ground of rejection. The claims which specify these and other process features provide advantages. It is respectfully submitted that these advantages would not have been obvious to one of skill in the art. Applicants, therefore, reserve the right to present further grounds of patentability based on one or more of these further features of the process of the invention. However, in view of the strong basis for patentability of the independent claims, and for claims specifying a difference in flow regime, it is respectfully submitted that this record need not be further extended by discussion of other novel and advantageous combinations that are defined by the various claims.

New Dependent Claims

New claims 200-247 have been added to claim the process in which used catalyst is contacted with additional N-(phosphonomethyl)iminodiacetic acid and oxygen for further production of glyphosate. The catalyst of the invention has been proven effective for extended continued use in a continuous process and repetitive use in a batch process, with relative modest deactivation.

Support for claims 200-247 may be found in the specification at p. 41, lines 26-27, p. 45, lines 13-22, p. 72, lines 1-18 and Table 12, and in Table 13, p. 77.

Double Patenting

Inasmuch as the instant application is a divisional application, presenting claims that were withdrawn subject to a restriction requirement in Ser. No. 09/248,655, it is respectfully submitted that the double patenting rejection is precluded by the provisions of 35 U.S.C. §121.

Although claims 100-199 are new, they are closely related to other claims directed to oxidation of N-(phosphonomethyl)glycine diacetic acid which were withdrawn subject to the restriction requirement in the parent case and presented here.

Applicants further note that, since both this application and Ser. No. 09/248,655 claim the same priority date, it is improbable that the term of a patent issuing on the instant application will extend significantly (if at all) beyond the term of a patent issuing on the '655 application. This consideration should further obviate the need for submission of such disclaimer. Nonetheless, based on the same consideration, should other issues be resolved without effect on the term, Applicants would agree to submission of a terminal disclaimer if double patenting should stand as the only barrier to allowance.

Information Disclosure Statement

The Examiner's attention is invited to the attached Third Supplemental Information Disclosure Statement. As noted above, this Statement provides information on commercial batch processes for the oxidation of N-(phosphonomethyl)iminodiacetic acid to glyphosate as practiced by Applicants' assignee more than a year prior to the priority date of the instant application. It further provides information on: a continuous process proposed by a third party for the oxidation of N-(phosphonomethyl)-iminodiacetic acid to glyphosate; and data obtained by Applicants regarding the CO desorption characteristics of a catalyst as prepared in accordance with Kim et al. *J. Electrochem. Soc.*, Vol. 140, No. 1, pp. 31-36; and data obtained by Applicants regarding the CO desorption and surface C/O ratio of a carbon only catalyst prepared in accordance with Chou U.S. Patents 4,624,937 and 4,696,772; a copy of Nitrokemia EPO patent 0 019 445; and information relating to Monsanto Company's opposition to the Nitrokemia EPO patent and its Australian analog.

For the reasons discussed in the Third Supplemental Information Disclosure Statement, it is respectfully submitted that the information provided therein does not constitute any obstacle to the patentability of the instantly claimed processes under 35 U.S.C. §103.

MARKED-UP VERSION SHOWING CHANGES

1. (once amended) A process for the preparation of N-(phosphonomethyl)glycine or a salt thereof, the process comprising contacting N-(phosphonomethyl)iminodiacetic acid or a salt thereof with an oxidation catalyst in the presence of oxygen, wherein the catalyst:

comprises a carbon support having a noble metal at a surface of the carbon support; and

is characterized as yielding no more than about 1.2 mmole of carbon monoxide per gram of catalyst when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant following the heating in the hydrogen atmosphere, is heated in a helium atmosphere [is heated] from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes.

13. (once amended) A process for the preparation of N-(phosphonomethyl)glycine or a salt thereof, the process comprising contacting N-(phosphonomethyl)iminodiacetic acid or a salt thereof with an oxidation catalyst in the presence of oxygen, wherein the catalyst comprises a carbon support having a noble metal, carbon, and oxygen at a surface of the carbon support[,];

said catalyst being characterized as having a [the] ratio of carbon atoms to oxygen atoms of at least about 20:1 at the surface [being at least about 20:1] as measured by x-ray photoelectron spectroscopy after the catalyst is heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before the catalyst is exposed to an oxidant following the heating in the hydrogen atmosphere.

23. (once amended) A process for the preparation of N-(phosphonomethyl)glycine or a salt thereof, the process comprising contacting N-(phosphonomethyl)iminodiacetic acid or a salt thereof with an oxidation catalyst in the presence of oxygen, wherein the catalyst comprises a carbon support comprising: (a) a noble metal at a surface of the carbon support; and (b) a surface layer having a thickness of about 50 Å as measured inwardly from the surface and comprising carbon and oxygen, the ratio of carbon atoms to oxygen atoms in the surface layer being at least about 20:1 as measured by x-ray photoelectron spectroscopy after the catalyst is heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before the catalyst is exposed to an oxidant following heating in the hydrogen atmosphere.

150. (once amended) A process as set forth in claim 147 wherein, before said oxidation of said reagent, the catalyst is characterized such that no more than about 1.2 mmole of carbon monoxide per gram of catalyst is desorbed when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant after following heating in a hydrogen atmosphere is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes.

151. (once amended) A process as set forth in claim 150 wherein, before said oxidation of said reagent, the catalyst is characterized such that no more than about 0.7 mmole of carbon monoxide per gram of catalyst is desorbed when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant after following heating in a hydrogen atmosphere is heated in a helium atmosphere from about 20 to

about 900°C at a rate of about 10°C per minute, and then at about
10 900°C for about 30 minutes.

152. (once amended) A process as set forth in claim 151
wherein, before said oxidation of said reagent, the catalyst is
characterized such that no more than about 0.5 mmole of carbon
monoxide per gram of catalyst is desorbed when a dry sample of
5 the catalyst, after being heated at a temperature of about 500°C
for about 1 hour in a hydrogen atmosphere and before being
exposed to an oxidant after following heating in a hydrogen
atmosphere is heated in a helium atmosphere from about 20 to
about 900°C at a rate of about 10°C per minute, and then at about
10 900°C for about 30 minutes.

153. (once amended) A process as set forth in claim 152
wherein, before said oxidation of said reagent, the catalyst is
characterized such that no more than about 0.3 mmoles of carbon
monoxide per gram of catalyst is desorbed when a dry sample of
5 the catalyst, after being heated at a temperature of about 500°C
for about 1 hour in a hydrogen atmosphere and before being
exposed to an oxidant after following heating in a hydrogen
atmosphere is heated in a helium atmosphere from about 20 to
about 900°C at a rate of about 10°C per minute, and then at about
10 900°C for about 30 minutes.

155. (once amended) A process as set forth in claim 154
wherein:

the catalyst further comprises carbon and oxygen at the
surface of the carbon support and before said oxidation of said
5 reagent, the catalyst is characterized as having a ratio of
carbon atoms to oxygen atoms of at least about 20:1 at the
surface of the carbon support as measured by x-ray photoelectron
spectroscopy after the catalyst is heated at a temperature of
about 500°C for about 1 hour in a hydrogen atmosphere and before

- 10 the catalyst is exposed to an oxidant following the heating in the hydrogen atmosphere.

Claims 200-247 are new.

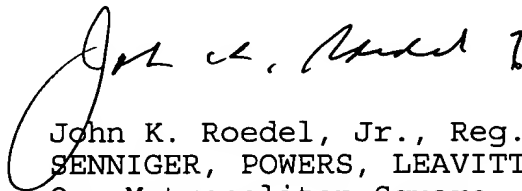
CONCLUSION

15 In view of the paucity of relevant teachings provided by
Franz, Hess, Hershman, Coloma and/or Ebner with respect to the
salient features of the process of the invention, and the
problems with which the claimed inventions deals, it is
respectfully submitted that claims 1-199 are not rendered obvious
by any combination of these references.

20 Favorable reconsideration and early allowance of all claims
are respectfully solicited.

A check in the amount of \$1,934.00 is enclosed to cover the fee
specified in 37 CFR §1.17(p) for submission of this Information
Disclosure Statement, a three-month extension of time and
25 additional claims. The Commissioner is hereby authorized to
charge any additional fees required under 37 CFR 1.16 and 1.17 or
refund any overpayment to Deposit Account No. 19-1345.

Respectfully submitted,



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